Local Atomic Structure in R-Mg-Zn, (R = Y, Gd, Dy and Tb)

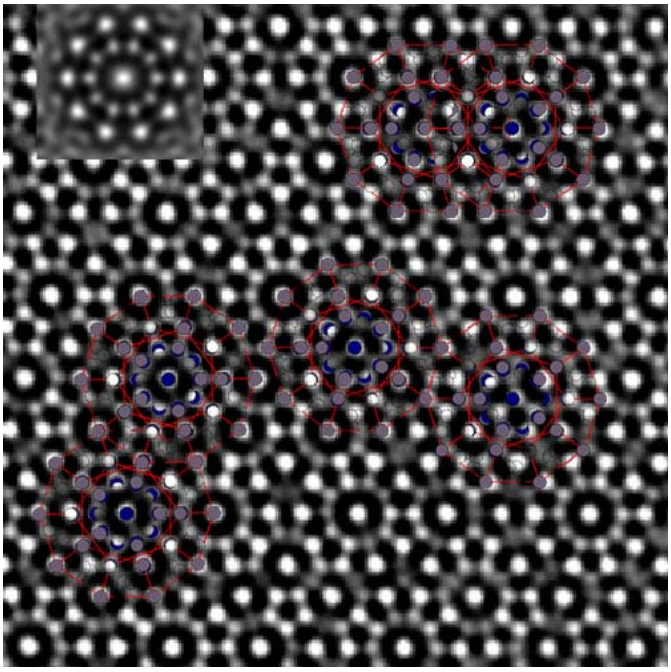
Personnel: M.J. Kramer (PI), Ya-Qiao Wu (Post-Doctoral Associate)

Abstract:

The aim of this study is to develop structural models of repetitive groupings or 'clusters' of atoms which are characteristic to each class of quasicrystals and their crystalline approximants. Despite their aperiodic order, these complex intermetallic compounds exhibit local atomic structures observed in chemically related crystalline compounds. The large differences in scattering cross-section between atoms in the rare-earth (R) containing compounds, their intrinsic magnetism and the intrinsically more tractable binary compounds lends the R class of quasicrystalline materials to a more thorough structural analysis than the Al-based quasicrystals.

Recent Results:

The large atomic number difference between Y and the heavy rare-earth (R) elements affords large differences in the scattering cross-sections for electrons, neutrons and X-rays. Using high-quality, flux-grown single grains with resolution-limited diffraction peaks; the local atomic structure was probed with a variety of complementary techniques. The differences in the atomic pair distribution function (PDF) due to loss of the anti-ferromagnetic ordering of the Tb between 4.2 and 30 K was used to probe the rare-earth site. High resolution TEM and single grain XRD have been used to determine the cluster structures. Results of this work suggest that there are local clusters in this face-centered icosahedral (fci) compound consisting of 5 concentric polyhedra containing 124 atoms. The proposed structure is similar to previous work and contains nested: icosahedron, dodecahedron, icosahedron, bucky-ball and a dodecahedron (see Table next page). However, the HRTEM results indicate that the local structure consists only of these 5 shells which may edge share with the next cluster. Based on this atomic arrangement, the PDF results are consistent with R atoms partially occupying the vertices of the outer icosahedron and dodecahedron.



High resolution TEM micrograph of the 5-fold axis shows aperiodic translation symmetry with 5 fold rotational symmetry. The overlays show edge sharing polyhedra clusters described above with different projections of the edge sharing along the 5 fold axis. In the upper left hand corner, a multi-slice calculation of a single cluster shows excellent correlation with the lattice image.

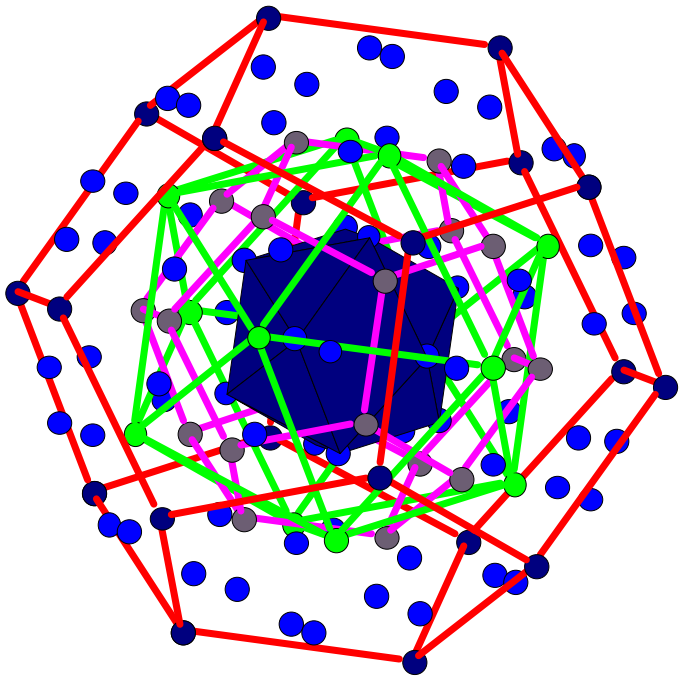
Significance:

Understanding the unique structure of quasicrystals and what gives rise to the aperiodic structure has been an open question since quasicrystals were first discovered by Dan Shechtman in 1982. Many of the Al-based quasicrystals are based on

Mackay or Bergman clusters. Whether the rare-earth quasicrystals are based upon any cluster model has been openly debated and remains uncertain. We believe that our work shows that the face centered icosahedral quasicrystals are based upon Bergman-like clusters and that the rare-earth provides a unique probe to determine the atomic structure.

Future Work:

While the atomic positions have been identified in the ternary R-Mg-Zn compound, the occupants of those sites have not been uniquely determined. The complexity of performing site occupation determinations in a binary compound is considerably reduced compared to the ternary. For this reason, efforts will shift to a newly discovered class of binary compounds that form primitive icosahedral (pi) quasicrystals, in close collaboration with Tom Lograsso of Ames Laboratory and Ian Fisher of Stanford University. To complement the structural studies, additional work on the composition, size and density of icosahedral clusters in the liquid prior to solidification will be preformed using high energy synchrotron radiation. The rational for this work is to provide insight into the role short range order plays a role in controlling the stability of quasicrystalline compounds.



Schematic drawing of the local cluster model proposed for the R-Mg-Zn fci compound. The R atoms preferentially occupy the outer icosahedron (green) and outer dodecahedron (dark blue) giving rise to a 8.7 Å separation between the anti-ferromagetically ordered rare earths. The length scales for these shells are given in the Table on the right.

Shell	Center to Vertex (Å)	Vertex Length (Å)
icosahedron	2.95	3.1
dodecahedron	4.68	3.34
icosahedron	5.53	5.53
C-60	6.85	2.65
dodecahedron	7.63	5.49

Interactions:

This broad interdisciplinary interaction includes Paul Canfield (Iowa State University and Ames Laboratory, Condensed Matter Physics) and Ian Fisher (Stanford University) on crystal growth, S.T. Hong and J.D. Corbett (Ames Laboratory, Materials Chemistry) on single grain XRD analysis and Y Zhu of Brookhaven National Laboratory assisting in the High Resolution Transmission Electron Microscopy.